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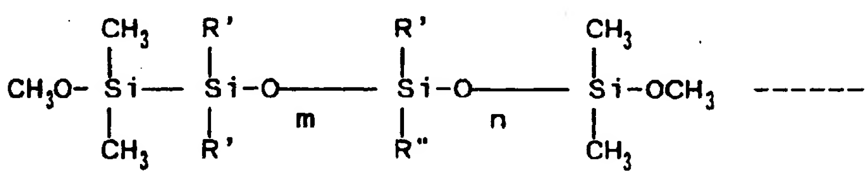
(54) **Polyester films with good release and slip properties and method for their preparation.**

(57) A method for preparation of a biaxially stretched polyester film with good slip and release properties comprising coating an acrylic resin-based aqueous resin compound, which is derived from adding an amino-modified silicone compound having the structural formula (1), a waxy additive and inert inorganic particles to an acrylic resin, on at least one surface of a mono-axially stretched polyester film, drying the polyester film coated with the aqueous resin compound, mono-axially stretching the dried polyester film in a direction perpendicular to that of the previous mono-axial stretching and heat-treating the stretched polyester film :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wherein R′ is a hydroxyl group, a methyl or an ethyl ; R″ is a hydrogen carbide which has 0 to 10 carbon atoms and to which NH or NH_2 is bonded ; m is an integer in the range of 5 to 1,000 ; n is an integer in the range of 100 to 20,000 ; and — 0.5.

A polyester film notably obtainable by the above method.

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BACKGROUND OF THE INVENTION

Field of the Invention

The present invention relates in general to a method for preparation of polyester films, and more particularly to a method for preparing biaxially stretched polyester films with high level and excellent release and slip properties by applying thereon with a surface coating layer.

Description of the Prior Art

As well known to those skilled in the art, known polyester films, particularly biaxially stretched polyester films, have excellent intrinsic properties, such as optical, mechanical and electrical properties, dimensional stability, heat resistance and chemical resistance. Thus, they are recently widely used for graphic art films, photo films, magnetic tape films, electrical insulation films, packing films and etc..

However, there occur several problems in processes, such as a printing process, a laminating process, a coating process and the like, for preparation of the several types of polyester film products using the known polyester film. For example, scratches may be easily generated on the film surface due to rolls with which the polyester film comes into close contact during the processes. Furthermore in this case, the higher the processing velocity increases the more scratches are generated. In addition, hot and humid atmosphere of the processes for preparing the film product detrimentally affects the slip property of the polyester film and this introduces several problems in working performances, for example, run ability of the polyester film and wind ablity of the film product, to the polyester film.

On the other hand, when the known polyester film is used for a release material in a fiberglass reinforced plastics (FRP) molding process, the release property of the release material is reduced at the interface between the FRP resin and the release material. Furthermore, the FRP curing conditions influence the release effect of the release material. Therefore, the release material may partially adhere to the FRP resin and, moreover, the sheet molding resin may adhere to the release material as it is heated.

In result, the known polyester films used for the release films in the FRP forming process have a problem in that they, particularly when they are very thin, may easily tear and/or be delaminated during the mechanical taking-up process thereof, thus causing the forming product to be damaged and, in this respect, to be inevitably reduced in its quality.

Furthermore, this problem of the known polyester film particularly happens when one polyester film is repeatedly used several times. Thus, it is requested to provide a polyester film which is capable of maintaining its own slip and release properties irrespective of repeated use

To impart excellent slip and release properties to the polyester film, it is preferred to apply, a silicone resin on at least one surface of the polyester film as well known to the art.

Conventionally, the silicone resin is applied to the polyester film surface by thermally curing the silicon resin in the presence of a curing agent. However, this silicone resin intrinsically has an excellent release property so that it appears deficient adhesion with respect to the base film. Additionally, if the silicone resin coating layer is strongly rubbed, it may be partially delaminated so as to be cloudy and, moreover, to be scaled off. Here, the tackiness of the silicone resin layer with respect to the base film is gradually reduced as time goes by and the reduction of the tackiness of the silicone resin layer is accelerated when the interface between the silicone resin layer and the base film surface is exposed to the atmosphere.

SUMMARY OF THE INVENTION

It is, therefore, an object of the present invention to provide a method for preparation of a polyester film having high level and excellent release and slip properties in which the above problems can be overcome and which comprises coating an acrylic resin-cased aqueous resin compound, which is derived from adding an amino-modified silicone compound, a waxy additive and inert inorganic particles to an acrylic resin, on at least one surface of a mono-axially stretched polyester film, drying the polyester film coated with the aqueous resin compound, mono-axially stretching the dried polyester film in a direction perpendicular to that of the previous mono-axial stretching and heat-treating the stretched polyester film.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

Another object of the present invention invention is to provide a new polyester film notably obtainable by the above process.

Preferably, the acrylic resin used in the present invention is a hard thermosetting emulsion resin, a thermoplastic water-dispersible resin and mixtures thereof. Here, each of the emulsion and water-dispersible resins comprises alkyl methacrylate and alkyl acrylate and has a glass transition temperature more than 40 °C.

In accordance with the improvement of this invention, the acrylic resin preferably comprises 40 to 90 molar % of alkyl methacrylate and/or alkyl acrylate and 10 to 60 molar % of vinyl monomer having a functional group capable of copolymerizing with the alkyl methacrylmate and/or the alkyl acrylate. When the acrylic resin contains the alkyl methacrylate and/or the alkyl acrylate not less than 40 molar %, it appears excellent blocking resistance and excellent coating layer forming property. On the other hand, when the acrylic resin contains the alkyl methacrylate and/or alkyl acrylate not more than 90 molar %, it easily permits a compound, which has a specified functional group as a copolymerization component, to be added thereto, thus causing the resultant acrylic resin to be easily dissolved in water and water-dispersed. Furthermore, such a dissolved and water-dispersed state of the acrylic resin is stably-maintained for a long time. In addition, such an acrylic resin exhibits excellent water resistance, chemical resistance and coating layer strength.

Also, the vinyl monomer, which has the functional group capable of copolymerizing with the alkyl methacrylate and alkyl acrylate, causes the resultant acrylic resin to have good hydrophile and high degree of water-dispersion and improves the stickiness between the polyester film base and the coating layer of the acrylic resin.

The alkyl group of the alkyl acrylate and the alkyl methacrylate is selected from methyl, ethyl, n-propyl, n-butyl, isopropyl, isobutyl, lauryl and etc..

The functional group, which is capable of copolymerizing with the alkyl methacrylate and the alkyl acrylate, is selected from a carboxyl group or a salt thereof, a sulfonic acid group or a salt thereof, an amide group or an alkylolated amide, a hydroxide, an epoxide and etc.

In view of the slip property, the coating layer strength, the water resistance and the chemical resistance, it is preferred to use a hard acrylic resin having a glass transition temperature not less than 40 °C.

The amino-modified silicone compound used in this invention has the structural formula:

$$CH_3O-\overset{\displaystyle CH_3}{\underset{\displaystyle CH_3}{\overset{|}{\underset{|}{Si}}}}-\Big(\overset{\displaystyle R'}{\underset{\displaystyle R'}{\overset{|}{\underset{|}{Si}}}}-O\Big)_m-\Big(\overset{\displaystyle R'}{\underset{\displaystyle R''}{\overset{|}{\underset{|}{Si}}}}-O\Big)_n-\overset{\displaystyle CH_3}{\underset{\displaystyle CH_3}{\overset{|}{\underset{|}{Si}}}}-OCH_3 \qquad \text{------} \quad (1)$$

wherein,

R′ is a hydroxyl group, a methyl or an ethyl,

R″ is a hydrogen carbide which has 0 to 10 carbon atoms and to which NH or NH_2 is bonded,

m is an integer in the range of 5 to 1,000,

n is an integer in the range of 100 to 20,000, and

$n/(m+n)$ [illegible] 0.5.

As described above, the amino-modified silicone compound has to have NH or NH_2 in its structural formula. The NH or NH_2 of the silicone compound reacts with the functional group, such as the carboxyl group, the sulfonic group, the hydroxyl group, the alkylol group, the epoxy group and etc., of the acrylic resin and causes the silicone compound to be prevented from being delaminated from the polyester film base.

To achieve good result or this invention, it is preferred to use an amino-modified silicone compound which is maintained in an emulsion or aqueous solution state and has a viscosity in the range of 5 to 500 cps. at a temperature of 25 °C. If the viscosity is lower than the above range, the slip and release properties of the resultant polyester film is detrimentally affected, while if the viscosity is higher than the above range, the compatibility between the amino-modified silicone compound and the acrylic resin is degraded.

On the other hand, the waxy additive is selected from polyolefin waxes having a plurality of structural formulas, preferably a polyethylene wax and a polypropylene wax. Here, the molecular weight of the used waxy additive is not particularly limited, but is preferably ranged from about 500 to about 50,000.

The inert inorganic particle is selected from titanium oxide, silicone oxide, calcium carbonate, kaoline, alumina, barium sulfate, zirconium, mica, calcium phosphate and etc., and has an average particle size in the range of about 20 to 1,000 nm, preferably in the range of about 50 to 300 nm.

In addition, it is preferred to mix required agents, such as a surface active agent, an antifoaming agent, a thickener, an antioxidant an ultraviolet absorber, a pH controller and etc. to the aqueous resin compound used

in this invention.

The used polyester film has a surface roughness not more than 0.03 μm, preferably not more than 0.01 μm, on the basis of the center line roughness measuring value and has a turbidity not more than 5.0, preferably not more than 1.0. Such a level, transparent polyester film is prepared by adding no organic or inorganic particle to a polyester resin or by adding a very small amount of the particles to the polyester resin. In addition, the film thickness is in the range of about 30 to 500 μm.

In this invention, the aqueous resin compound is coated on at least one surface of the polyester film base by a conventional coating method prior to accomplishment of the crystal orientation of the polyester film base. In other words, the aqueous resin compound is applied to the polyester film base just after mono-axial stretching of the melted and extruded film base, followed by drying in a conventional drying process. Thereafter, the polyester film is stretched in a direction perpendicular to the prior stretching direction and in turn heat treated.

This aqueous resin compound has a solid content not more than 20 % by weight, preferably not more than 8 % by weight. Also, the viscosity of the aqueous resin compound is not more than about 100 cps., preferably not more than about 20 cps..

As described above, the coating method for coating the above aqueous resin compound on the polyester film base is selected from the conventional coating methods, such as direct roll coating, reverse roll coating, direct gravure coating, reverse gravure coating, bar coating and the like. Here, the coating amount of the aqueous resin compound is in the range of 0.001 g. to 1 g. per ft^2 of the final product, preferably in the range of 0.005 g. to 0.2 g. per ft^2 of the final product.

The polyester film products prepared by the method of this invention exhibit excellent slip and release properties so that they can be wide used for micro films, labels and various types of laminate products.

In this invention, the characteristics of the polyester film products are measured as follows:

1. Coefficient of Friction

The coefficients of statical friction μ_s and the coefficients of kinetic friction μ_K of the final products were measured using a slippery measuring instrument, which was manufactured by Dongyang Tester Co. of Korea, under a load of 200 g and in accordance with ASTM D1894.

2. Release Property

A) Pink lacquer method:

To measure the release properties of the polyester film products, each polyester film product, on which the aqueous resin compound had been coated, was coated with a pink lacquer as a testing resin, followed by drying using a conventional drier. The film product was then formed with a plurality of lattice slits spaced apart from each other by a distance of 1 mm. A scotch tape (Model No. 610 manufactured and sold by 3M Co.) as an adhesive tape then strongly adhered to the lattice slit surface of the film product with no air bag formed therebetween. Thereafter, the scotch tape was suddenly took off the film surface and the number of lattices, each having the area of 1 × 1 mm^2 and being delaminated from the polyester film surface due to the aforementioned sudden taking off of the tape, was calculated in terms of percentage.

B) Printing ink method:

To measure the release properties of the polyester film products, each polyester film product, on which the aqueous resin compound had been coated, was coated with an aqueous gravure ink as a testing ink and dried by a conventional drier. Thereafter, the same procedure as that of the aforementioned pink lacquer method was repeated. The number of lattices was calculated in terms of percentage.

3. Surface Consistency (Center Line Average Roughness)

The average surface consistencies of the polyester film products were measured using a consistometer SE-3H (manufactured and sold by KOSAKA Laboratory of Japan) from hanged charts under the condition of 2 μm of radius of the needle, 30 mg of load, 20,000 magnifications, 0.08 mm of cut-off value.

The following examples and comparative examples are merely intended to illustrate the present invention in further detail and should by no means be considered to limitative of the scope of the invention.

EXAMPLE 1

Acrylic resin (A): a thermosetting acrylic emulsion resin which comprised 50:50 molar ratio of alkyl acrylate and alkyl methacrylate and was mixed with an anionic surface active agent having a glass transition temperature of 50 °C.

Acrylic resin (B): a thermoplastic acrylic water-dispersible resin which comprised alkyl acrylate and alkyl methacrylate of molar ratio in the range of 50:50 to 99:1 and was mixed with an anionic surface active agent having a glass transition temperature of 100 °C.

Polyethylene terephthalate, which had an inherent viscosity of 0.65 when it was measured in the presence of 25 °C of orthochlorophenol, was melted and extruded, thereafter, the extrusion was rapidly cooled on a drum maintained at a temperature of 40 °C so as to produce a non-orientated film having a thickness of 650 μm. The resultant film was then mono-axially stretched using a metal roll, which is maintained at a temperature of 95 °C, to 3.6 times in the running direction of the film. Thereafter, the stretched film was coated with an aqueous resin compound derived from mixing 65 % by weight of the acrylic resin (A), 29 % by weight of the amino-modified silicone compound, 3 % by weight of the waxy additive and 3 % by weight of the inert inorganic particles, followed by drying. The dried film was then stretched to 3.6 times in a direction perpendicular to the running direction of the film at a temperature of 105 °C and heat-treated at a temperature of 220 °C, thereby preparing a biaxially stretched polyester film product having 50 μm thickness and 0.02 g/m^2 of coating amount of the coating layer. The characteristics of the polyester film product are given in Table 1.

EXAMPLE 2

The procedure of Example 1 was repeated, substituting 68 % by weight of the acrylic resin (A) for 65 % by weight of the resin (A) and using no waxy additive. The characteristics of the biaxially stretched polyester film product are given in Table 1.

EXAMPLE 3

The procedure of Example 1 was repeated, substituting 68 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A), 25 % by weight of the amino-modified silicone compound for 29 % by weight of the compound, 3.5 % by weight of the waxy additive for 3 % by weight of the wax and 3.5 % by weight of the inert inorganic particles for 3 % by weight of the particles, respectively. The characteristics of the biaxially stretched polyester film product are given in Table 1.

EXAMPLE 4

The procedure of Example 1 was repeated, substituting 60 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A) and 34 % by weight of the amino-modified silicone compound for 29 % by weight of the compound, respectively. The characteristics of the biaxially stretched polyester film product are given in Table 1.

EXAMPLE 5

The procedure of Example 1 was repeated, substituting 78 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A), 15 % by weight of the amino-modified silicone compound for 29 % by weight of the compound and 4 % by weight of the inert inorganic particles for 3 % by weight of the particles, respectively. The characteristics of the biaxially stretched polyester film product are given in Table 1.

EXAMPLE 6

The procedure of Example 1 was repeated, substituting 13 % by weight of the acrylic resin (A) and 52 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A), 26 % by weight of the amino-modified silicone compound for 29 % by weight of the compound, 5 % by weight of the waxy additive for 3 % by weight of the wax and 4 % by weight of the inert inorganic particles for 3 % by weight of the particles, respectively. The characteristics of the biaxially stretched polyester film product are given in Table 1.

EXAMPLE 7

The procedure of Example 1 was repeated, substituting 33 % by weight of the acrylic resin (A) and 33 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A), 26 % by weight of the amino-modified silicone compound for 29 % by weight of the compound, 4 % by weight of the waxy additive for 3 % by weight of the wax and 4 % by weight of the inert inorganic particles for 3 % by weight of the particles, respectively. The characteristics of the biaxially stretched polyester film product are given in Table 1.

EXAMPLE 8

The procedure of Example 1 was repeated, substituting 85 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A), 7 % by weight of the amino-modified silicone compound for 29 % by weight of the compound, 4 % by weight of the waxy additive for 3 % by weight of the wax and 4.5 % by weight of the inert inorganic particles for 3 % by weight of the particles, respectively. The characteristics of the biaxially stretched polyester film product are given in Table 1.

EXAMPLE 9

The procedure of Example 1 was repeated, substituting 89 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A), 2 % by weight of the amino-modified silicone compound for 29 % by weight of the compound, 4.5 % by weight of the waxy additive for 3 % by weight of the wax and 4.5 % by weight of the inert inorganic particles for 3 % by weight of the particles, respectively. The characteristics of the biaxially stretched polyester film product are given in Table 1

EXAMPLE 10

The procedure of Example 1 was repeated, substituting 68 % by weight of the acrylic resin (B) for 65 % by weight of the acrylic resin (A) and 29 % by weight of the amino-modified silicone compound for 29 % by weight of the compound, respectively, and using no waxy additive. The characteristics of the biaxially stretched polyester film product are given in Table 1.

COMPARATIVE EXAMPLE 1

The procedure of Example 1 was repeated, coating no aqueous resin compound on the polyester film. The characteristics of the biaxially stretched polyester film product are given in Table 1.

COMPARATIVE EXAMPLE 2

The procedure of Example 1 was repeated, substituting an aqueous resin compound comprising 100 % by weight of the acrylic resin (A) for the aqueous resin compound. The characteristics of the biaxially stretched polyester film product are given in Table 1.

COMPARATIVE EXAMPLE 3

The procedure of Comparative Example 2 was repeated, substituting an aqueous resin compound comprising 100 % by weight of the acrylic resin (B) for the aqueous resin compound. The characteristics of the biaxially stretched polyester film product are given in Table 1.

COMPARATIVE EXAMPLE 4

The procedure of Comparative Example 2 was repeated, substituting an aqueous resin compound comprising 50 % by weight of the acrylic resin (A) and 50 % by weight of the acrylic resin (B) for the aqueous resin compound. The characteristics of the biaxially stretched polyester film product are given in Table 1.

TABLE 1

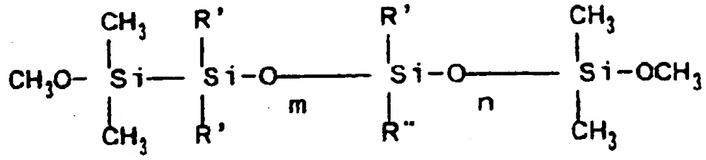
Content / Section	Coefficient* μ_s	μ_k	Release Property Pink Lacquer	Printing Ink	Surface Consistency
Exam. 1	0.28	0.23	100	100	0.007
Exam. 2	0.41	0.36	100	100	0.007
Exam. 3	0.25	0.22	60	95	0.007
Exam. 4	0.27	0.20	100	100	0.007
Exam. 5	0.38	0.29	40	80	0.008
Exam. 6	0.27	0.25	100	100	0.007
Exam. 7	0.27	0.24	100	100	0.007
Exam. 8	0.47	0.38	0	20	0.007
Exam. 9	0.50	0.43	0	3	0.008
Exam. 10	0.44	0.37	100	100	0.007
Comp. 1	1.0↑	1.0↑	0	0	0.007
Comp. 2	0.56	0.47	0	0	0.009
Comp. 3	0.59	0.48	0	0	0.009
Comp. 4	0.57	0.48	0	0	0.009

*: Coefficient of friction

The present invention has been described in detail with particular reference to preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit and scope of the invention.

Claims

1. A method for preparation of a biaxially stretched polyester film with good slip and release properties comprising the steps of:
   - coating with at least one surface of a mono-axially stretched polyester film; an acrylic resin-based aqueous resin compound, which is derived from adding an amino-modified silicone compound having the structural formula (1),

 ------ (1)

wherein

R′ is a hydroxyl group, a methyl or an ethyl;

R″ is a hydrogen carbide which has 0 to 10 carbon atoms and to which NH or NH_2 is bonded;

m is an integer in the range of 5 to 1,000;

n is an integer in the range of 100 to 20,000; and

n/(m+n) == 0.5.

a waxy additive and inert inorganic particles to an acrylic resin,

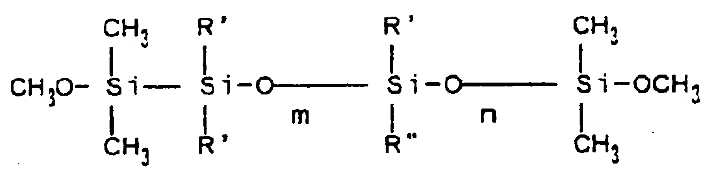
- drying the polyester film coated with said aqueous resin compound;
- mono-axially stretching the dried polyester film in a direction perpendicular to that of the previous mono-axial stretching; and
- heat-treating the stretched polyester film.

2. The method according to claim 1, characterized in that said acrylic resin is selected from a hard thermosetting emulsion resin, a thermoplastic water-dispersible resin and mixtures thereof, said thermosetting emulsion resin and thermoplastic water-dispersible resin comprising alkyl methacrylate and alkyl acrylate and having a glass transition temperature not less than 40°C, respectively.

3. The method according to one of claims 1 or 2, characterized in that said amino-modified silicone compound amounts in the range of 5 to 75% by weight of solid concentration of said acrylic resin.

4. The method according to one of claims 1 to 3, characterized in that said waxy additive is a polyolefin wax having a molecular weight in the range of 500 to 50,000.

5. A biaxially stretched polyester film notably obtainable by a method comprising the steps of:
   - coating with at least one surface of a mono-axially stretched polyester film; an acrylic resin-based aqueous resin compound, which is derived from adding an amino-modified silicone compound having the structural formula (1),

 ------ (1)

wherein

R′ is a hydroxyl group, a methyl or an ethyl;

R″ is a hydrogen carbide which has 0 to 10 carbon atoms and to which NH or NH_2 is bonded;

m is an integer in the range of 5 to 1.000;

n is an integer in the range of 100 to 20,000; and

n/(m+n) == 0.5.

- a waxy additive and inert inorganic particles to an acrylic resin,
- drying the polyester film coated with said aqueous resin compound;
- mono-axially stretching the dried polyester film in a direction perpendicular to that of the previous mono-axial stretching; and
- heat-treating the stretched polyester film.

6. The polyester film according to claim 5, characterized in that said acrylic resin is selected from a hard thermosetting emulsion resin, a thermoplastic water-dispersible resin and mixtures thereof, said thermo-

setting emuslion resin and thermoplastic water-dispersible resin comprising alkyl methacrylate and alkyl acrylate and having a glass transition temperature not less than 40°C, respectively.

7. The polyester film according to claim 5 or 6, characterized in that modified silicone compound amounts in the range of 5 to 75% by weight of solid concentration of said acrylic resin.

8. The polyester film accorrding to one of claims 5 to 7, characterized in that said waxy additive is a polyolefin wax having a molecular weight in the range of 500 to 50,000.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 92 40 2209

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	US-A-4 098 840 (M.YOSHIDA ET AL.) * column 7, line 48 - column 8, line 6; claim 9 * ---	1,5	C08J7/04 C09D133/06 //(C09D133/06, 183:08) C08L67:02
A	EP-A-0 189 978 (UNITED MERCHANTS AND MANUFACTURERS, INC.) * claims 5,7,8 * ---	1,5	
A	EP-A-0 359 017 (HOECHST CELANESE CORPORATION) * claims 1-10 * ---	1	
A	EP-A-0 039 225 (AMERICAN HOECHST CORPORATION) * claims 1,9 * -----	1,2,5,6	

TECHNICAL FIELDS SEARCHED (Int. Cl.5)

C08J
C09D

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	12 NOVEMBER 1992	HALLEMEESCH A.D

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document